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# Enhancement of a Semi-Batch Chemical Reactor Efficiency through its Dimensions Optimization

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**Abstract**

Efficiency of manufacturing processes is very important in today's competitive world with hard economic rules. In chemical engineering area the efficiency depends on the production heart, which is often a chemical reactor. In this paper authors describe process of optimal semi-batch exothermic reactor dimensions finding. The task is to find reactor dimensions which lead to process efficiency improving, i.e. to processing the greater chemicals amount in the same or shorter time. The optimizing process uses an objective function which includes also the reactor mathematical model. The objective function is modified here to find better results and necessary limitations and penalizations are used to improve the dimensions searching. An evolutionary approach represented by the Self-Organizing Migrating Algorithm (SOMA) was used to minimize the defined function. A suitability of the reactor with newly found dimensions was then verified by the process control simulations.



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*Keywords:* Temperature control; Online identification; Semi-batch reactor; Exothermic process; SOMA

## 1. Introduction

Even if the problem of the chemical reactors efficiency is quite important and is connected with the reactor dimensions, there are not many studies published about the reactor dimensions optimization these days. Usually, the process is placed to existing vessel and then the authors try to control the process with different control techniques. But, the process is often not suitable for the given reactor geometry. Here, the authors are trying different way – to suite the reactor geometry to the exact process – to achieve the best control results.

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Some published papers which describe an effect of a reactor size on the process control are mentioned further. The „performance“ of three bubble column reactors with the same volume but varied geometry (height to diameter ratios – 0.93; 2.04; 3.98) was investigated in [1]. It was demonstrated, that larger height-to-diameter ratio could enhance $CO_2$ capture efficiency. Paper [2] tries to observe more efficient membrane reactor setups in order to improve production yield. Optimization has been carried out by a differential evolution considering 40 decision variables including dimensions of membrane reactor. The authors concluded that the obtained variables improved the „performance“ of the continuous catalyst regeneration reformer process. Park et al. [3] developed a 2D mathematical model of a pilot-scale fixed-bed reactor for a Fe-based Fischer-Tropsch synthesis. The developed model clearly showed the effects of the tube diameter on the temperature profiles, in terms of the radial heat transfer area. Varga et al. [4] tried to find optimal feeding profile of fed-batch chemical reactor. The optimal feeding profile was generated with sequential quadratic programming, classical evolutionary strategy and the advanced version of evolutionary strategy based on covariance matrix adaptation. Any publications dealing with a semi-batch reactor dimensions optimization to compare it with here presented results were not found unfortunately.

Our paper is organized as follows: In section 2, the semi-batch reactor, its structure, mathematical model and the process control are described; section 3 presents simulation results and section 4 concludes the current work and suggests new areas for investigation.

## 2. Methods section

### *2.1. Current situation*

In past different control techniques have been applied on fed-batch reactor presented in this paper [5,6,7]. The goal was to optimize the reactor temperature using two manipulated variables (the reactor feeding and the cooling water temperature) to achieve the shortest possible one batch cycle time.

**Nomenclature**

| | |
|---|---|
| $A$ [$s^{-1}$] | Pre-exponential factor |
| $a_{FK}(t)$ [-] | Mass concentration of the chromium sludge |
| $c_{FK}$ [$J.kg^{-1}.K^{-1}$] | Chromium sludge specific heat capacity |
| $c_R$ [$J.kg^{-1}.K^{-1}$] | Reactor content specific heat capacity |
| $c_v$ [$J.kg^{-1}.K^{-1}$] | Coolant specific heat capacity |
| $E$ [$J.mol^{-1}$] | Activation energy |
| $\Delta H_r$ [$J.kg^{-1}$] | Reaction heat |
| $K$ [$J.m^{-2}.K^{-1}.s^{-1}$] | Conduction coefficient |
| $k$ [$s^{-1}$] | The reaction rate constant |
| $m(t)$ [kg] | Weight of the reaction components in the system |
| $\dot{m}_{FK}$ [$kg.s^{-1}$] | Mass flow of the entering chromium sludge |
| $\dot{m}_v$ [$kg.s^{-1}$] | Coolant mass flow |
| $m_{vR}$ [kg] | Coolant mass weight in the reactor double wall |
| $R$ [ $J.mol^{-1}.K^{-1}$] | Gas constant |
| $S$ [$m^2$] | Heat transfer surface |
| $T(t)$ [K] | Temperature of reaction components in the reactor |
| $T_{FK}$ [K] | Chromium sludge temperature |
| $T_v(t)$ [K] | Temperature of coolant in the reactor double wall |
| $T_{vp}$ [K] | Input coolant temperature |

Here authors describe other achievements obtained by further optimization. The aim was to optimize the reactor dimensions, i.e. the radius and the height. Default dimensions of the original reactor were: radius: 0.78 m, height: 1.11 m, volume: 2.12 $m^3$, surface: 7.35 $m^2$. The initial values used by simulations follow: flow of the cooling water – 4.67 $kg.s^{-1}$, temperature of the cooling water – 274.58 K, temperature of the filter cake – 293.15 K, temperature inside the reactor – 323.15 K.

*2.2. The mathematical model*

The fed-batch reactor process optimization is performed on a data obtained from a real chromium sludge regeneration process. This process is described in previous work [7].

The leather industry is a producer of pollutants in the form of chrome-tanned solid waste. This waste is a potential threat to human health, because it contains trivalent chromium (Cr III), which can under various conditions oxidize to its hexavalent form (Cr VI). One of the numerous possible solutions of this problem is the chrome-tanned waste enzymatic dechromation [9]. The whole process is supposed to run in the reactor located in Kortan company in Hrádek nad Nisou [10]. Because of that fact the mathematical model was designed for this reactor, which dimensions and geometry are shown in the Fig. 1.

To simulate tanning salts from the chromium sludge regeneration process a mathematical model is used. The reactor has four input signals $\dot{m}_{FK}(t)$, $\dot{m}_V(t)$, $T_{FK}(t)$, $T_{VP}(t)$ and one output signal $T(t)$ (see the Table 1.). The chemical reactor scheme is shown in Fig. 2.

The fed-batch reactor mathematical model is defined by four differential equations - Eq. (1-4).

$$\dot{m}_{FK} = \frac{d}{dt} m(t) \tag{1}$$

$$\dot{m}_{FK} = k\, m(t)\, a_{FK}(t) + \frac{d}{dt}\left[m(t) a_{FK}(t)\right] \tag{2}$$

$$\dot{m}_{FK} c_{FK} T_{FK} + \Delta H_r k\, m(t)\, a_{FK}(t) = K\, S\left[T(t) - T_V(t)\right] + \frac{d}{dt}\left[m(t)\, c_R\, T(t)\right] \tag{3}$$

$$\dot{m}_V c_V T_{VP} + K\, S\left[T(t) - T_V(t)\right] = \dot{m}_V c_V T_V(t) + m_{VR} c_V\, T_V'(t) \tag{4}$$

The first equation expresses the total mass balance of chemicals in the reactor. The symbol $\dot{m}_{FK}$ [$kg.s^{-1}$] expresses the mass flow of entering chromium sludge, $\frac{d}{dt} m(t)$ the accumulation of the in-reactor content.

The second equation expresses the chromium sludge mass balance. The input is $\dot{m}_{FK}$ [$kg.s^{-1}$] again, the accumulation is $m(t)\frac{d}{dt} a_{FK}(t)$ and the express $k\, m[t] a_{FK}[t]$ means the chromium sludge extinction by the chemical reaction. Symbol k[$s^{-1}$] means reaction rate constant expressed by an Arrhenius equation (5).

$$k = A e^{-\frac{E}{RT[t]}} \tag{5}$$

The Arrhenius equation parameters are important for the process control, especially for the process safety as is described in [11]. All reactor variables and parameters are described in nomenclature part on the previous page.

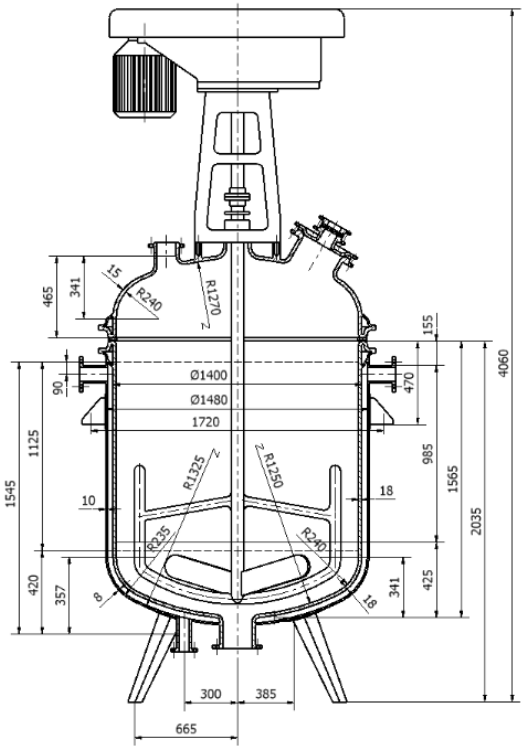


Fig. 1. Exothermic chemical semi-batch reactor.

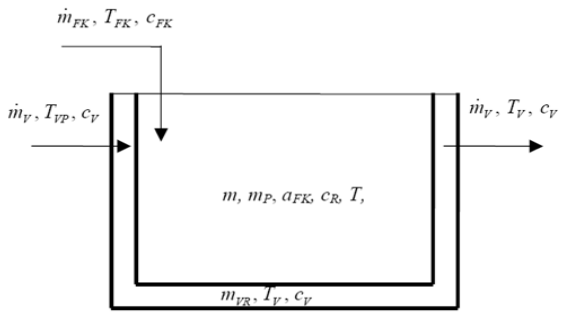


Fig. 2. Chemical reactor scheme.

The third equation describes enthalpy balance. The reactor input heat is expressed by $\dot{m}_{FK} c_{FK} T_{FK}$, the heat rising from the chemical reaction is $\Delta H_r k\, m[t] a_{FK}[t]$, the reactor wall heat transmission is $K S (T[t] - T_V[t])]$ and the accumulated heat inside the reactor is described by $m(t) c_R \frac{d}{dt} T(t)$. The last equation describes a coolant balance. The coolant input heat is expressed by, $\dot{m}_V c_V T_{VP}$ the heat entering the coolant from the reactor by the reactor wall is $K S (T[t] - T_V[t])$, the heat going out with the coolant is $\dot{m}_V c_V T_V[t]$ and the heat accumulated in the double wall

coolant expresses $m_{VR}c_V \frac{d}{dt}T_V(t)$. This model cannot be solved by analytical means. It is necessary to use numerical methods – the Matlab and Mathematica software were used for this task.

### *2.3. SOMA (Self-Organizing Migrating Algorithm)*

An objective function was defined for the reactor dimensions optimization. As was said before, the mathematical model has to be solved by numerical means. Because of it, also the objective function has to be solved numerically. To find the objective function minimum a SOMA algorithm was used.

SOMA is a stochastic optimization algorithm that is based on the social behavior of cooperating individuals. It can be also classified as an evolutionary algorithm, despite the fact that no new generations of individuals are created during the search. Only the positions of the individuals in the search space are changed during a generation (also called a 'migration loop'). Individuals are generated randomly according to what is called 'specimen of the individual' principle [12]. SOMA algorithm was chosen for its ability to converge towards the global minimum.

This algorithm tried to find three optimal variables: radius of the reactor, height of the reactor and feeding of chromium sludge. There is no requirement of the constant volume in contrast to the mentioned work [1]. The only limits are minimum and maximum radius <0.1 m, 6 m>, height <0.1 m, 6 m> and feeding <0.01 kg.s$^{-1}$, 3 kg.s$^{-1}$>.

The main input variables of SOMA were set as follows: **Step** - indicates the size of a migration step, was chosen 0.3, **PRT** - perturbation parameter modifies the movement vector of an individual to the leader (0.1), **NP** - number of individuals in population was equaled 50, **Migrations** - number of migration cycles was 10, **AcceptedError** - the maximum allowed difference between the best and the worst individual in the population (0.1).

### *2.4. Objective function*

Objective function helps to find the reactor with required properties. This function was formed gradually. Basic formula (Eq. 6) is used to find minimum of the difference $w - T[t]$, where $w$ is the desired and $T[t]$ is the current in-reactor temperature. The formula in (Eq. 6) will be referred as OF 1.

$$f_{OF} = \sum_{0}^{t} |w - T[t]| \tag{6}$$

The desired in-reactor temperature value was *370 K*. Processing of the chromium sludge is *faster* at *higher* temperature, i.e. close to the desired value. For safety reasons the temperature should not exceed *370 K*.

Using this objective function SOMA algorithm found new dimensions of the reactor: *radius = 1.428 m*; *height = 1.044 m*. The reactor with these new dimensions values was then used to simulate the control of whole process. The simulation was finished when the in-reactor temperature dropped to the default temperature *323.15 K*. The time to process one batch took *24850 s* (Fig. 3).

The amount of the processed chromium sludge obtained from the first simulation was *2019 kg* (Fig. 3. Filling up the reactor). It is actually the difference between the weight of the initial filing and the weight of the final batch, i.e. *7729-5710=2019 kg*.

The reactor performance was calculated from the two above mentioned values - the batch time and the processed chromium sludge weight amount. Its value was *0.081 kg.s$^{-1}$*. The value was further divided by coefficient *3.15* to be compared with the original reactor whose volume is *3.15* times smaller. The final performance of newly designed reactor was then *0.0258 kg.s$^{-1}$*. The new reactor dimensions caused a small performance improvement, as the original reactor performance was *0.0237 kg.s$^{-1}$*.

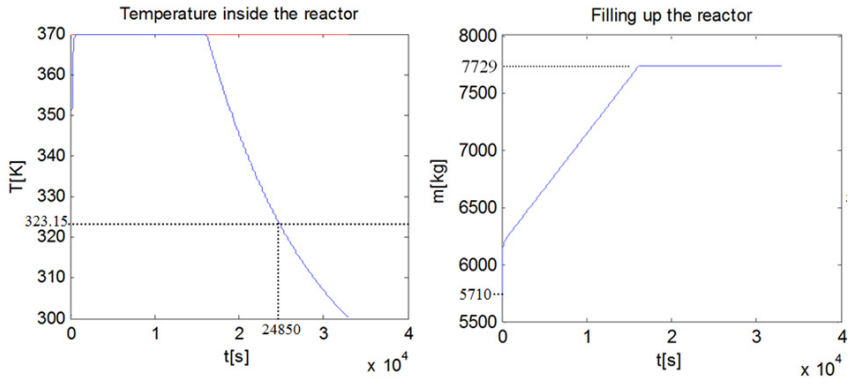


Fig. 3. The mixture temperature and weight inside the reactor, OF 1.

The further results obtained by the reactor control simulation are showed in Fig. 4. and Fig. 5. In the Fig. 4. can be seen the coolant temperature development and the actuating signal (the reactor feeding) changes caused by the controller. The coolant temperature has the similar course as the in-reactor temperature and is not so important in this case. The actuating signal changes are important from the practical point – too many fast changes can significantly shorten the actuator working life. The concentration of the chromium sludge (Fig. 5.) is also important, chromium sludge accumulation is connected with the heat development delay, i.e. the temperature does not follow the concentration immediately as the chromium sludge is dosed because of the reaction kinetics.

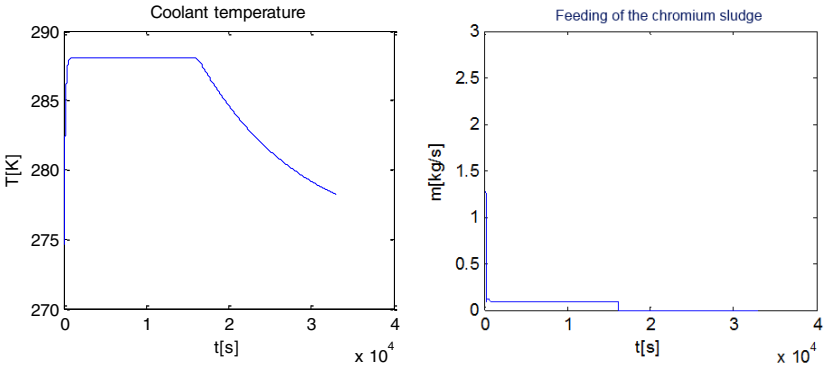


Fig. 4. Temperature of the cooling medium and the feeding mixture amount, OF 1.

To get even better results, the objective function was extended with the concentration term $\sum_{0}^{t} a_{FK}[t]$, see (Eq. 7):

$$f_{OF} = \sum_{0}^{t} |w - T[t]| + \sum_{0}^{t} a_{FK}[t] \tag{7}$$

The newly added term should allow faster decrease of concentration of chromium sludge which leads to faster completion of the process. This extended objective function is further referred as OF 2.The results obtained by simulation show that the hypothesis was right, but the impact was negligible. The reactor performance increased

only to the value *0.0288 kg.s*$^{-1}$. The performance was again calculated from the total process time (*22212 s*) and the amount of processed chromium sludge (*3110 kg*). The total time and the processed amount can be observed in graphs (Fig. 6.). Dimensions of the reactor here were: The reactor radius = *2.73 m*; height = *0.44 m*.

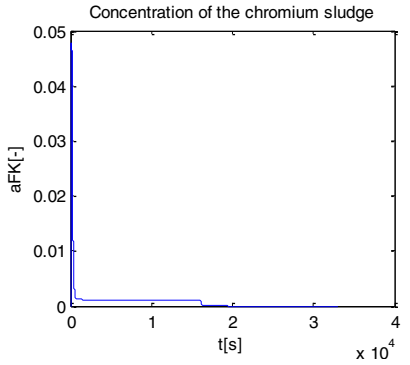


Fig. 5. Concentration of the mixture inside the reactor, OF 1.

The last change in objective function definition was done by multiplying the concentration term by 2 (Eq. 8). This last objective function is further referred as OF 3.

$$f_{OF} = \sum_{0}^{t} |w - T[t]| + 2\sum_{0}^{t} a_{FK}[t] \tag{8}$$

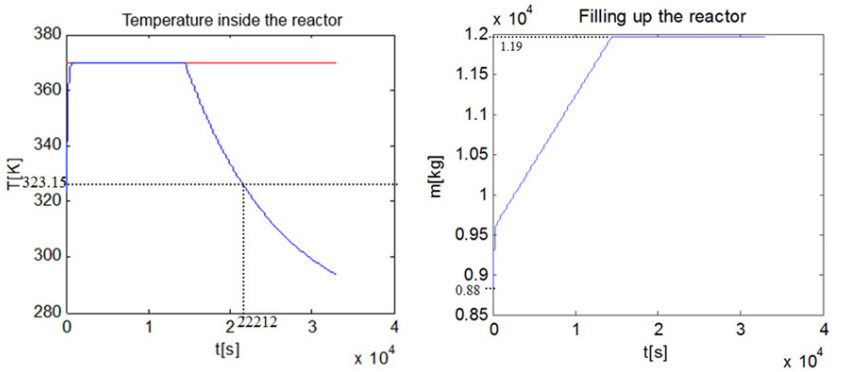


Fig. 6. Temperature of the cooling medium and the feeding mixture amount, OF 2.

The above mentioned change should increase the impact of the concentration term in the objective function and thus help to find better solution. The results placed in Fig. 7. validated the hypothesis again. Here the performance improved significantly, the new value was 0.0395 kg.s$^{-1}$. The total process time was in this case 16195 s and the amount of processed chromium sludge 2788 kg. The new dimensions: The reactor radius = 3.41 m; height = 0.25 m. The time and the processed amount are showed in Fig. 7.

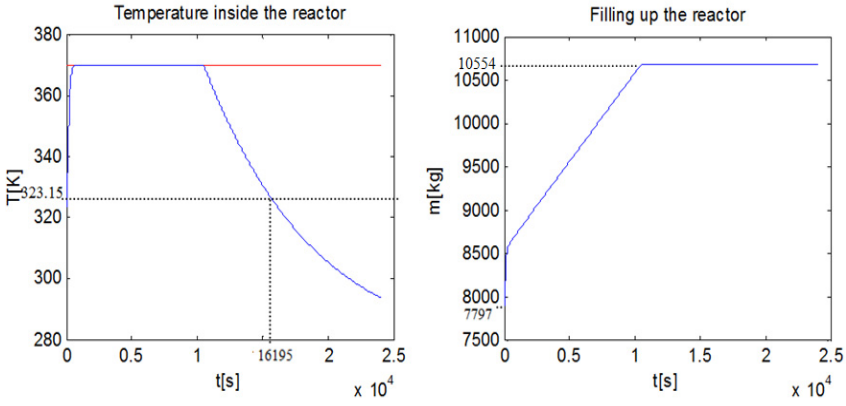


Fig. 7. Temperature of the cooling medium and the feeding mixture amount, OF 3.

In the last simulation step the task was to restrict the reactor volume near to specific value to prevent the reactor of being too small or large. To do so the interval in objective function was limited by penalization. The penalization is used in case of exceeding the minimum or the maximum volume. The minimum volume was set as 1 $m^3$, the maximum as 10 $m^3$. If the algorithm goes out of these values, the objective function is penalized by the 50000. This value was found experimentally. The performance 0.07 $kg.s^{-1}$ obtained here has the best value from all simulations round and increased the value almost 3 times, if compared to the original reactor (0.0237 $kg.s^{-1}$). The other values were: process time 9110 s, amount of processed chromium sludge 2408 kg, radius = 4.42 m; height = 0.13 m. The time and the processed amount can be found in Fig. 8.

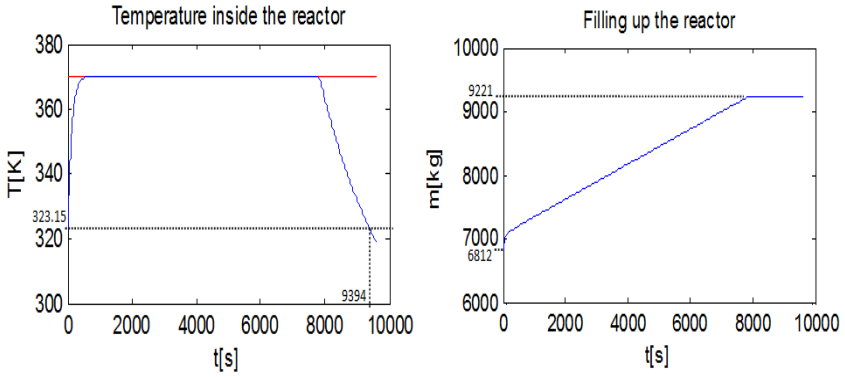


Fig. 8. Temperature of the cooling medium and the feeding mixture amount, penalization.

## 3. Results section

As was already said, the task was to find such reactor, whose dimensions fit our process better than the original one. Practically, it means that we would be able to process a larger quantity of material in a shorter time than it was possible with the original reactor. To do so, we were looking for such reactor diameter, height and input chemical dosing which fulfil best these requirements. We proceeded in the following manner – at first, the new reactor dimensions were found by the static optimization and some other corresponding data as the total in-reactor mass and the initial reactor filling were counted. Secondly, the newly founded reactor obtained by the static optimization was used for the process control simulation and was controlled by the Ziegler-Nichols controller to find the total process

time. From the processing time and the processed amount the reactor "performance" was counted. All results presented here were obtained after the successful control applied on the reactor optimized by the static optimization. The control is necessary here to keep the in-reactor temperature at the highest levels, because the process chemical reaction runs faster with higher temperature, and also to keep the process safe.

The values obtained by the simulations can be seen in Table 1. The first three columns contain values calculated by SOMA algorithm for the reactor dimensions static optimization. Fourth column contains the objective function value. The fifth column shows reactor „performance". The „performance" means how much of chromium sludge can reactor process per second. The „performance" is in relationship with the reactor volume. Practically, the „performance" means that if the new reactor has double volume, an obtained value of processed chromium sludge is multiplied by a coefficient 0.5 to compare it with the original volume reactor. Even if the simulations were done just as a first try, the calculations showed that the newly found reactor dimensions can improve our process efficiency. As can be seen from the results of simulations in the Table 1, all new reactors were with better "performance". From the „performance" point of view the original reactor shows the „performance" 0.0237 $kg.s^{-1}$, the new average value obtained by simulations was 0.0766 $kg.s^{-1}$ i.e. approximately 3 times higher.

Table 1. Values found by SOMA algorithm.

| Radius [m] | Height [m] | Feeding [$kg.s^{-1}$] | Objective Function [-] | Performance [$kg.s^{-1}$] | Volume [$m^3$] |
|---|---|---|---|---|---|
| 2.02116 | 0.117457 | 0.0897612 | 10571.7 | 0.091 | 1.5 |
| 0.590243 | 1.37818 | 0.0450906 | 16879.5 | 0.044 | 1.51 |
| 0.250407 | 1.69088 | 0.0200795 | 10512.3 | 0.092 | 0.33 |
| 2.51136 | 0.155479 | 0.132499 | 13454 | 0.063 | 3.17 |
| 2.68451 | 0.100654 | 0.141601 | 10605.3 | 0.093 | 2.29 |

## Conclusion

The task was to find such reactor dimensions which leads to better process efficiency. It practically means to process greater amount of inputs in the same or shorter time then it was with the original reactor. The optimizing process used an objective function which included also the reactor mathematical model. The objective function was modified gradually to find better results and necessary limitations and penalizations were used to improve the dimensions searching. An evolutionary approach represented by the Self-Organizing Migrating Algorithm (SOMA) was used to minimize the defined function. A suitability of the reactor with newly found dimensions was then verified by the process control simulations. The efficiency of a suggested reactor was compared with the original reactor previously used for the same process. It was found that the reactor geometry characterized by height and diameter plays an important role in the process efficiency. Based on the satisfactory control results and better reactor „performance", we can say that the new reactor dimensions are more suitable for our exothermic process. In future, the impact of height-to-width ratio will be investigated to find even more suitable variations for this process. The "winning" reactors described in this paper have the ratio reciprocal – first is low and wide, second high and slim.

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